1,162,436

PATENT SPECIFICATION

NO DRAWINGS

1,162,436

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Date of filing Complete Specification: 8 March, 1968.

Date of Application (No. 12815/67): 18 March, 1967.

Complete Specification Published: 27 August, 1969.

Index at acceptance:—C3 P(7A, 7D1B, 7T2X); C5 F(A, 477, 600, 605, 619, 678, 762, 808); C5 G1A1

International Classification:—C 08 f 27/08

COMPLETE SPECIFICATION

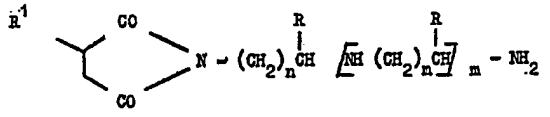
Ashless Dispersants

We, OROBIS LIMITED, of Devonshire House, Mayfair Place, London, W.1, a British Company, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to a process for making certain compounds and their use in lubricant compositions and fuels.

The compounds made by the process of the present invention find use as ashless dispersants, i.e. they will disperse fuel and lubricant oxidation products produced in an internal combustion engine and prevent aggregation of these particles, thus preventing the deposition of carbon and lacquer on engine parts. The compounds are therefore useful as fuel and lubricant additives.

Accordingly the present invention is a process for producing compounds suitable for use as fuel and lubricant additives by the reaction of a compound of formula



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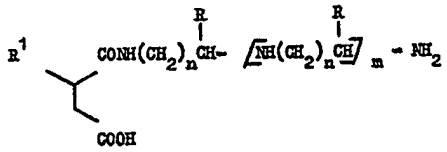
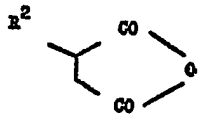


II

with a compound of formula



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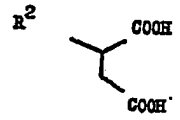


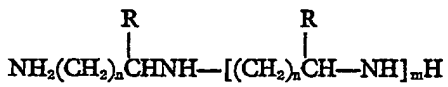
wherein n is zero or an integer and m is zero or an integer, R is hydrogen or an alkyl radical and R^1 and R^2 are susbtantially aliphatic hydrocarbon radicals in which either R^1 or R^2 has a chain length of less than 50 carbon atoms and the arithmetic mean of the chain lengths of R^1 and R^2 is greater than 50 carbon atoms. The integer n is preferably at least 1.

The compounds I and II may be prepared, for example by the reaction of an alkylene amine with a susbtantially aliphatic substituted derivative of succinic acid or succinic anhydride, for example a polyalkylene substituted acid or anhydride. It is

[*Price 4s. 6d.*]

preferred to react either equimolar quantities or an excess of the alkylene amine with the succinic acid or anhydride derivative, ratios of amine to succinic acid or anhydride derivative of 1:1 to 1:0.5 being suitable. The product of this reaction is a mixture whose exact composition depends on the reaction conditions employed, but which includes compounds I and II. The reaction requires heating, for example to a temperature between about 80°C and 200°C. It is preferred to use a temperature above 100°C to remove water produced during the reaction. The reaction may be carried out in the presence of a diluent oil or a solvent. Solvents which will form azeotropes with water, for example, toluene, are preferred. Nitrogen may also be blown through the mixture to expel water.

The alkylene amine is a compound of formula



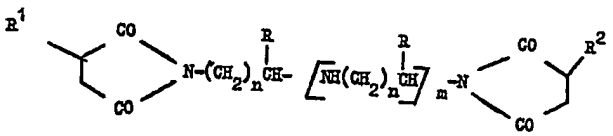
where n, m and R are as defined above. The radical may contain up to 6 carbon atoms and is preferably hydrogen or a methyl radical. Suitable compounds include ethylene diamine, when m is zero, n is 1 and R is hydrogen; 1,2-propylene diamine when m is zero, n is 1 and R is methyl; and 1,3-propylene diamine when m is zero, n is 2 and R is hydrogen. Other examples of suitable compounds are diethylene triamine, triethylene tetramine, tetraethylene pentamine, tri-1,2-propylene tetramine, tri-1,3-propylene tetramine and octaethylene nonamine, i.e. m may suitably have values up to 9.

The compound I or II, or a mixture of compounds I and II is reacted with a quantity of a substantially aliphatic substituted derivative of succinic acid or succinic anhydride to give the desired reaction product. The substantially aliphatic substituent is preferably a polyalkylene radical. The amounts of compound I and/or II and polyalkylene derivative of succinic acid or anhydride used are not critical. The molar ratio of compound I and/or II to succinic acid or anhydride derivatives used depends upon the amine employed but preferably lies in the range 0.5:1 to 2:1. The reaction may be carried out under conditions described above for the preparation of compounds I and II.

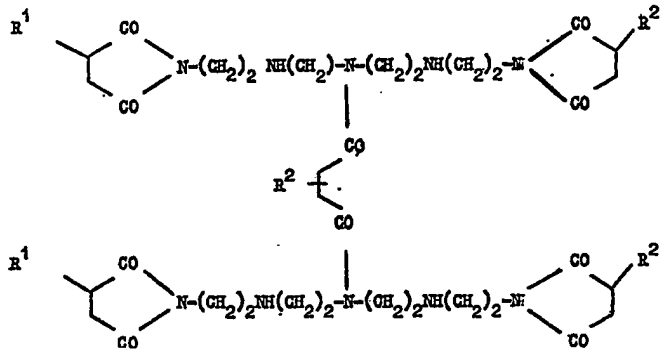
The substituted succinic acids and anhydrides used in these reactions may be obtained by the reaction of an olefin, alkyl chloride or alcohol with maleic anhydride. The compound which is reacted with maleic anhydride is frequently derived from polymers or copolymer s of, for example, ethylene, propylene or butylene. These compounds which are commercially available are not in fact single compounds of a definite chain length but mixtures of compounds, and the chain length quoted is the average chain length of the compounds in such a mixture. Where appropriate, reference to the chain length of a substituent is to be understood as reference to the average chain length of a mixture of substituents.

The chain length of either R^1 and R^2 is less than 50 carbon atoms, preferably in the range 12 to 49. The chain length of the other radical is greater than 50 and preferably less than 250 carbon atoms, the range being preferably 50 to 100. The length is sufficient to raise the arithmetic mean length of the chain lengths R^1 and R^2 to greater than 50. A radical having a chain length of 70 carbon atoms is particularly suitable for use in the process of this invention.

It is believed that the products obtained by the process of this invention have the following structure, but is not known with certainty.



In the case where the compound I or II contains more than one free amine group, for example when the alkylene amine used in its production is a polymer such as tetraethylene pentamine, and it is reacted with an excess of substituted succinic acid or anhydride, a compound whose structure is believed to be the following may be obtained.



The invention is illustrated in the following example.

EXAMPLE

Tetraethylene pentamine (150 g.) was warmed to 50°C and polyisobutyl succinic anhydride (Mwt 760, 47 carbon atoms in the side chain) as an oil solution (1614 g.) added slowly with vigorous agitation. The temperature was raised to 150°C. and a stream of nitrogen passed through the solution to void expelled water. The resultant intermediate was cooled to 100°C and polyisobutenyl succinic anhydride (Mwt = 1200, 78 carbon atoms in the side chain) in oil solution (1650 g.) added. The temperature was raised to 150°C and held at this temperature for 1 hour. To expel all the water of reaction the reactants were further heated to 180°C for 1 hr at reduced pressure. The product, a brown clear oil solution, had a nitrogen content of 1.6%.

The product of the Example was evaluated in the Petter A.V. 1 diesel engine run to Ministry of Defence Specification DEF 2101D. (Petter is a registered Trade Mark). The test blend consisted of the product of the Example (1.03%) + calcium phenate (29 mM) + zinc dialkyl dithiophosphate (8mM) in which the alkyl groups were derived from a mixture of C_4 and C_6 alcohols, in solvent Refined High VI Middle East Stocks blended to SAE 30.

By the symbol "mM" is meant millimoles of active metal present per kilogram of finished lubricant.

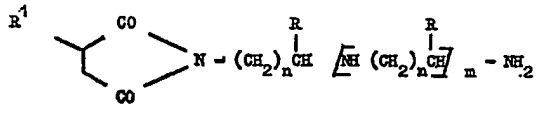
The result is given in the following table.

Component	Possible Rating	Actual Rating	Acceptable Limit
Groove 1	10	5.5	<5
2	10	8.5	"
3	10	7.0	"
4	10	8.7	"
Land 1	10	5.1	"
2	10	7.3	"
3	10	6.7	"
Average Ring groove and Land merit	10	6.9	7
Skirt Exterior	10	9.95	"
Skirt Interior	10	9.4	"
Overall Rating	80	59	>55

The Acceptable limit is a guide line issued by the Ministry of Defence.

WHAT WE CLAIM IS:—

1. A process for producing compounds suitable for use as fuel and lubricant additives by the reaction of a compound of formula



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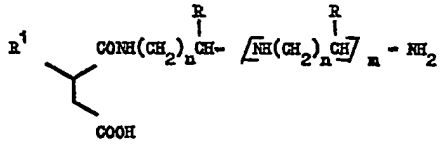
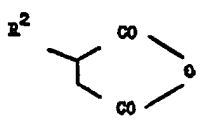


II

with a compound of formula



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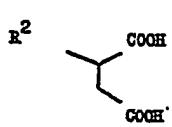

wherein n is zero or an integer and m is zero or an integer, R is hydrogen or an alkyl radical, R^1 and R^2 are substantially aliphatic hydrocarbon radicals in which either R^1 or R^2 has a chain length of less than 50 carbon atoms and the arithmetic mean of the chain length of R^1 and R^2 is greater than 50 carbon atoms.

2. A process according to claim 1 wherein the integer n has the value 1 or 2.

3. A process according to claims 1 or 2 wherein the integer m has the value 1 to 9.

4. A process according to any of the preceding claims wherein R is an alkyl radical having up to 6 carbon atoms.

5. A process according to claim 4 wherein R is a methyl radical.

6. A process according to any of the preceding claims wherein the compound of formula I or II is prepared by reaction of an alkylene amine with a polyalkylene substituted derivative of succinic acid or anhydride.

7. A process according to claim 6 wherein the alkylene amine is ethylene diamine, 1,2-propylene diamine, 1,3-propylene diamine, diethylene triamine, triethylene tetramine, tetraethylene pentamine, tri-1,2-propylene tetramine, tri-1,3-propylene tetramine or octaethylene nonamine.

8. A process according to claim 6 or 7 wherein the reaction is carried out at a temperature in the range 80°C to 200°C.

9. A process according to claim 8 wherein the reaction is carried out above 100°C.

10. A process according to any of claims 6 to 9 wherein the reaction is carried out in the presence of a solvent which will form an azeotrope with water.

11. A process according to claim 7 wherein the solvent is toluene.

12. A process according to any of the preceding claims wherein the molar ratio of compound I and/or II reacted with the substituted succinic acid or anhydride lies in the range 0.5 : 1 to 2 : 1.

13. A process according to claim 12 wherein the reaction is carried out at a temperature in the range 80° to 200°C.

14. A process according to claim 13 wherein the reaction is carried out above 100°C.

15. A process according to any of claims 12, 13 or 14 wherein the reaction is carried out in the presence of a solvent which will form an azeotrope with water.

16. A process according to claim 15 wherein the solvent is toluene.

17. A process according to any of the preceding claims wherein the chain length of either R^1 or R^2 is in the range 12 to 49 carbon atoms.

18. A process according to claim 17 wherein the chain length of the radical which is not in the range 12 to 49 carbon atoms is in the range 50 to 250 carbon atoms.

19. A process according to claim 18 wherein the chain length of the radical is in the range 50 to 100 carbon atoms.

20. A process for producing compounds suitable for use as fuel and lubricant additives substantially as herein described with particular reference to the Example.

21. Compounds produced by the process of any of the preceding claims.

22. Fuels or lubricants containing any of the compounds claimed in claim 21.

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Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1969.
Published by the Patent Office, 25 Southampton Buildings, London, W.C.2, from which copies may be obtained.